

High-resolution neutron-scattering study of the roton in confined superfluid ^4He

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We report high-resolution neutron inelastic-scattering measurements of the roton energy and linewidth of superfluid ^4He confined in 95% porosity aerogel glass for temperatures $0.08 < T < 1.2$ K. The confined roton line shape can be described well by a damped harmonic oscillator (DHO) function, as in the bulk. The energy of the confined roton is essentially temperature independent over this temperature range. The roton linewidth does not exhibit a finite width of greater than $0.1 \mu\text{eV}$ at low temperature. This result is in contrast to previous measurements carried out at lower resolution. [S0163-1829(99)01118-2]

INTRODUCTION

The excitation spectrum of bulk superfluid ^4He consists of a phononlike excitation at low-momentum transfer, $Q < 1 \text{ \AA}^{-1}$, and a minimum in energy at around $Q = 1.9 \text{ \AA}^{-1}$ known as the roton minimum. This very sharp roton excitation disappears completely at temperatures above the superfluid transition temperature T_λ , and has come to be regarded as a signature of the superfluid phase.^{1,2} At low temperatures, roton-roton interaction theory³ predicts that the width of the roton in the bulk liquid tends towards zero corresponding to an infinite lifetime. This has been shown to be in good agreement with experiment.⁴

Recent interest has focused on the effects of confinement of superfluid ^4He in porous glasses. Aerogel glass has an open-pore morphology and consists of a network of silica strands giving rise to a broad distribution of pore sizes (ranging from 50–500 Å). The high-resolution experiments of Dimeo *et al.*,⁵ Soko *et al.*,⁶ and Gibbs *et al.*⁷ showed that, while the low-temperature excitation energies of the confined liquid are very similar to those of the bulk, their variation with temperature is markedly different. When ^4He is confined in the pores of aerogel glass, some roton broadening may be expected due to the restricted geometry imposed by its pores. This broadening has been estimated by Gibbs

*et al.*⁷ to be of the order of $0.1 \mu\text{eV}$. However, at 1.3 K, Gibbs *et al.*⁷ reported a roton width of some $10 \mu\text{eV}$. Recently an extensive study by Plantevin *et al.*,⁸ mapping out the behavior of the low-energy excitations of ^4He confined in 95% porosity aerogel, reported a roton width of $6 \mu\text{eV}$ at 0.5 K.

The upgrade of the neutron backscattering spectrometer⁹ (IN10) at the Institut Laue-Langevin has allowed us to access the momentum and energy transfer region of interest with an inelastic energy resolution of $< 1 \mu\text{eV}$. This is a substantial improvement on those employed in all of the previous studies: Plantevin *et al.* used an inelastic resolution of $110 \mu\text{eV}$ and Dimeo *et al.*, Sokol *et al.*, and Gibbs *et al.* used an elastic resolution of $15 \mu\text{eV}$. We find that at the lowest temperature measured, the broadening of the excitation due to confinement in the aerogel pores does not exceed $0.1 \mu\text{eV}$.

EXPERIMENTAL DETAILS

The experiments were carried out on the inverse geometry spectrometer, IN10, at the Institut Laue-Langevin, Grenoble. The high energy resolution of IN10 (configuration IN10B) is obtained by orienting both the NaF(111) monochromator crystals and the Si(111) analyzer in a backscattering configuration with a final neutron energy of 2.08 meV. The incident

neutron energies are selected by Bragg scattering from the monochromator crystal. The energy scan through the region of interest is obtained by heating and cooling the monochromator, and hence varying the lattice parameter by thermal expansion. In order to correct for possible errors due to hysteresis with scan direction, two scans (one heating, and one cooling the monochromator) were performed for each (sample) temperature measured. To ensure that the correct momentum transfer Q was studied, it was necessary to improve the Q resolution, ΔQ , of IN10. This was achieved by restricting the exposed area of the analyzer crystals to the locus of $Q(\hbar\omega) = 1.935 \pm 0.005 \text{ \AA}^{-1}$ with a suitable cadmium mask.

The aerogel glass used in the measurement was of 95% porosity and was made by the decomposition of tetramethoxysilane solution into $\text{Si}(\text{OH})_4$ and methanol. $\text{Si}(\text{OH})_4$ is unstable and polymerizes readily into colloidal aggregates of SiO_2 releasing water. The result is a silica glass mesh immersed in methanol. Removal of the methanol is performed by a hypercritical drying process. The helium sample was condensed into a cylindrical cell containing 13 cm^3 of the glass. The intensity of the observed roton peak corresponded well to the open pore volume of the aerogel, when compared to previous measurements on IN10.¹⁰ Roton measurements were made at 77 mK, and then at six temperatures between 800 and 1200 mK.

DATA ANALYSIS

Extracting detailed information on the location and width of the roton peak requires knowledge of the peak shape and instrumental resolution. It is customary to use a low-temperature measurement of the bulk liquid to model the instrumental resolution, since at low temperature the roton width is expected to be extremely small.³ Because the IN10 monochromator crystal is changed between experiments, the resolution measurement is unique to each experiment. Such a low-temperature bulk measurement was not available for this experiment. However, Fig. 1 compares the observed scattering from ^4He confined in aerogel at $T = 77 \text{ mK}$ at a momentum transfer of $Q = 1.93 \text{ \AA}^{-1}$ with the corresponding data for bulk ^4He obtained on the same instrument (Andersen *et al.*).⁴ Both scans are asymmetric with a shoulder on the side of greater neutron energy loss. This is caused by the combination of the imperfect Q resolution of IN10 and the dispersion of the helium excitations. The intrinsic instrumental energy resolution is close to Lorentzian; however the (approximately quadratic) dispersion of the rotons over the ΔQ leads to an asymmetric resolution function. The extent of the asymmetry is well known, since it is directly related to the width of the cadmium mask defining the ΔQ of IN10 (see Andersen *et al.*⁴ for further details). The width of the Lorentzian was found to be 0.8 \mu eV in the aerogel and 0.7 \mu eV in the bulk. A broadening of this magnitude is consistent with the limitations imposed by the repeatability of the instrumental setup and shows that the broadening of the roton signal, brought about by confinement in aerogel does not exceed 0.1 \mu eV at low temperatures. Therefore we used the low-temperature aerogel scans to model the instrumental resolution for this experiment. In the fitting procedure the momentum transfer Q_R and the effective mass μ_R of the roton were

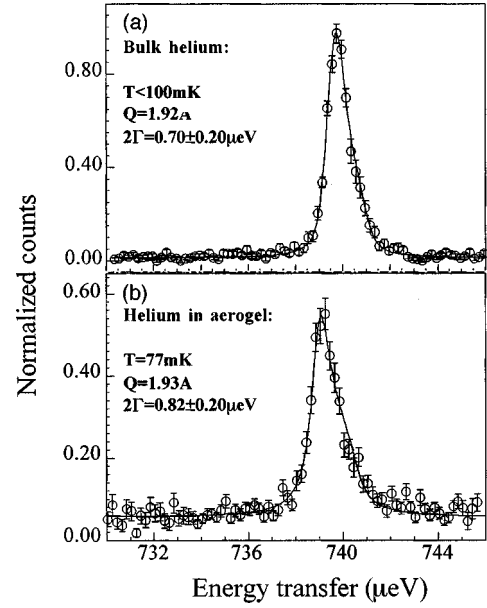


FIG. 1. Raw data (open circles) and least-squares fits to the model described in the text (solid line) at the lowest temperatures measured for (a) bulk ^4He , $T < 100 \text{ mK}$ (Ref. 4), (b) ^4He in aerogel, $T = 77 \text{ mK}$, this work.

fixed at 1.92 \AA^{-1} and 0.14 , respectively, whereas the roton energy, peak height, peak width, and flat background were left to vary as free parameters.

The observed line shape in bulk helium can be described well by a damped harmonic oscillator (DHO) (Ref. 11) function:

$$S(Q, \omega) = \frac{Z(Q)}{\pi} [n_B(\omega) + 1] \times \frac{4\omega\omega_Q\Gamma_Q}{[\omega^2 - (\omega_Q^2 + \Gamma_Q^2)]^2 + (2\omega\Gamma_Q)^2}, \quad (1)$$

where $n_B(\omega)$ is the Boltzmann distribution function and $Z(Q)$, ω , ω_Q , and Γ_Q are the strength, energy transfer, energy, and width of the roton excitation, respectively. This function can also be used to describe the observed scattering of ^4He in aerogel. For the higher temperature scans, the data at temperatures from 0.85 – 1.0 K and from 1.05 – 1.15 K were binned into channels of 0.2 and 0.4 \mu eV , respectively, and then were fitted by least squares to the convolution of the DHO function with the resolution function discussed above. The background was found to be independent of temperature and was held fixed at its 77 mK value; the height, width, and position of the DHO function were left to vary as free parameters.

RESULTS AND DISCUSSION

The fitted values of peak position ω_Q and width Γ_Q are shown in Table I and Fig. 2. The widths of the confined roton are very similar to those of the bulk and the energy is seen to be temperature independent over this temperature range. At the highest temperatures, the sizes of the error bars are significantly larger. This is because a separate analysis of the scans of the same temperature but different scan direction

TABLE I. Values of peak width Γ and energy ω .

Temperature (K)	2Γ (μeV)	ω (μeV)
0.85	0.46 ± 0.14	739.1 ± 0.1
0.90	0.73 ± 0.11	739.2 ± 0.2
0.95	0.86 ± 0.28	739.1 ± 0.3
1.00	1.73 ± 0.51	739.1 ± 0.2
1.05	1.86 ± 0.54	739.0 ± 0.1
1.10	2.83 ± 1.08	739.40 ± 0.2
1.15	3.75 ± 1.48	739.11 ± 1.1

(temperature of the monochromator) showed that there was a marked difference between those scans taken while the monochromator was being heated and those taken while it was being cooled. Hence the absolute differences between the values of the energy and linewidth of each have been added to the statistical error bars. The systematic errors arising from the instrumental effects are expected to be negligible for the width of the roton, but of the order of $5 \mu\text{eV}$ for the *absolute* excitation energy.

The temperature dependence of the linewidth of the roton excitation is very well described by a model first proposed by Landau and Khalatnikov¹² and later refined by Bedell, Pines, and Zawadowski³ (BPZ). Two rotons, one created by an incoming neutron and the other excited thermally, combine and then decay into two other quasiparticles. This leads to a dependence on temperature of the linewidth $\Gamma_Q(T)$ given by Eq. (2)

$$\Gamma_Q(T) = 3.585(1 + 0.0603T^{1/2})T^{1/2} \exp[-\omega_Q(T)/k_B T]. \quad (2)$$

This model predicts that the width of the roton tends towards zero at low temperatures corresponding to an infinite lifetime at 0 K. Agreement between measured widths and the BPZ calculations are excellent for bulk ^4He .⁴ However, the temperature dependence of the energy of the bulk roton is not yet fully understood, with recent measurements⁴ showing a clear departure of behavior from that predicted by the theory of Bedell *et al.* below 1.3 K.

There has been much speculation surrounding the behavior of the lifetime of the confined roton at low temperatures. Gibbs *et al.*⁷ showed that a decrease in the roton lifetime due to collisions with aerogel pore walls would correspond to an excitation broadening of the order of only $0.1 \mu\text{eV}$. These authors also performed a Monte Carlo simulation which indicated that multiple scattering should not give rise to any significant broadening of the roton excitation. It is very important therefore to establish how the roton does behave at low temperatures and, in particular, whether it exhibits a finite value at low temperatures since a “saturation” of the width would be an indication that other decay processes occur. The recent measurements of Plantevin *et al.* on aerogel of 95% porosity⁸ indicated that a finite roton width of 6

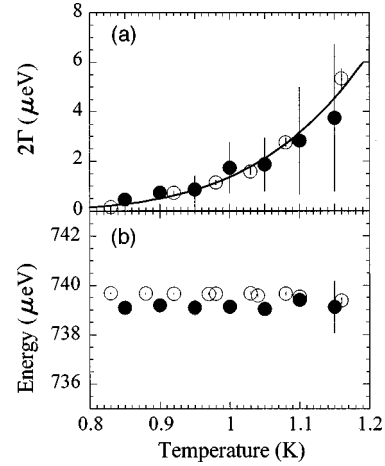


FIG. 2. Variation of (a) the roton linewidth and (b) roton energy gap as a function of temperature for helium in aerogel (solid circles) and bulk helium (open circles) (Ref. 4). Solid line in (a) is the temperature dependence of the roton according to the BPZ theory (Ref. 3).

$\pm 1 \mu\text{eV}$ remains at 500 mK. Gibbs *et al.*⁷ found a roton width of some $10 \mu\text{eV}$, but at the relatively elevated temperature of 1.3 K. The measurements reported here were performed with substantially better resolution than the previous studies (full width at half maximum $< 1 \mu\text{eV}$ in these measurements compared with full width at half maximum $= 110 \mu\text{eV}$)⁸ and show quite clearly that any broadening at the lowest temperature does not exceed $0.1 \mu\text{eV}$. The disagreement possibly arises from the differences in instrumental energy resolution. We note that resolving a scattering feature of width $6 \mu\text{eV}$ with a resolution function of width $110 \mu\text{eV}$ results in an observed linewidth very little different from the underlying resolution width. It is also possible that what manifests itself as broadening of the peak in the Plantevin data arises from additional scattering around the roton energy. Very broad peaks would be observable in the Plantevin data, but appear only as a small background component in our data. It is impossible to confirm the presence of anomalous extra background in our data since the background contains contributions from the aerogel and adsorbed impurities.

In summary, neutron experiments with energy resolution of less than $1 \mu\text{eV}$ show that any low-temperature broadening of the roton excitation due to the confinement of superfluid ^4He in porous aerogel glass does not exceed $0.1 \mu\text{eV}$. We plan to continue these investigations to study the effect of different pore geometries on the roton excitations.

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